FACULTY OF SCIENCES

EPR STUDY OF LUMINESCENCE-BASED RADIATION DOSIMETERS

K. Maes[1], L. Lowie[1], J. Kusakovskij[1], L. de F. Nascimento[2], H. Vrielinck[1], F. Callens[1]

[1]GHENT UNIVERSITY DEPARTMENT OF SOLID STATE SCIENCES, EMR RESEARCH GROUP

[2]SCK•CEN BELGIAN NUCLEAR RESEARCH CENTRE, RADIATION PROTECTION DOSIMETRY AND CALIBRATION GROUP

Introduction

Electron paramagnetic resonance (EPR) is a physical method to observe resonant absorption of microwave radiation by unpaired electron spins in a magnetic field. It is able to detect, identify and quantify free radicals, such as those present in irradiated materials. This makes it a reliable dosimetric technique for retrospective/accident dosimetry, detection of irradiated food, e.g. using alanine, tooth enamel and sucrose. It finds applications in geology, chemistry, physics, medicine, environmental sciences, archaeology, and industrial irradiations. [1] In the present study, we discuss results on two classes of materials, i.e. LiF:Mg,Ti /LiF:Mg,Cu,P and Al_2O_3:C /Al_2O_3:C,Mg. These materials are used in thermoluminescence dosimetry (TLD) and Optically Stimulated Luminescence dosimetry (OSLD)/Radiophotoluminescence dosimetry (RPLD) respectively.[2-4] In terms of dose sensitivity EPR cannot compete with these luminescence methods, but it can provide complementary insight into the defects and processes leading to luminescence.

Al_2O_3:C /Al_2O_3:C,Mg

X-Band measurements

Both systems were measured before and after X-ray irradiation. Before irradiation, there was no significant EPR signal visible. After irradiation a broad isotropic signal appeared. Although the signal looks similar for both, they are in fact slightly different:

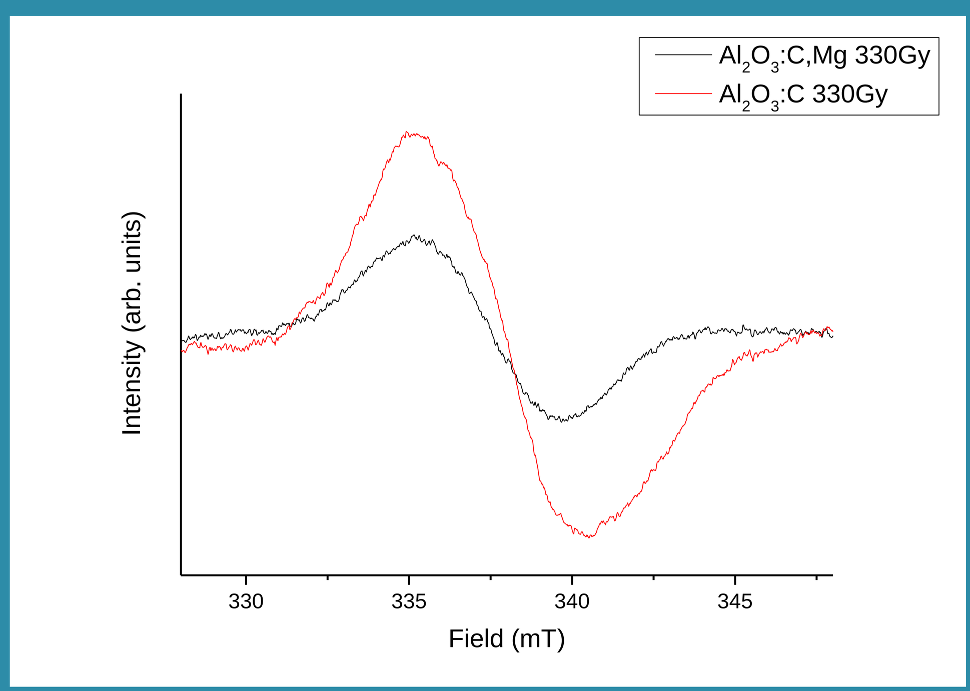
Al_2O_3:C,Mg

g = 2,011 lwpp = 4,3 mT

Al_2O_3:C

g = 2,008 lwpp = 5,3 mT

X-band EPR comparison, RT



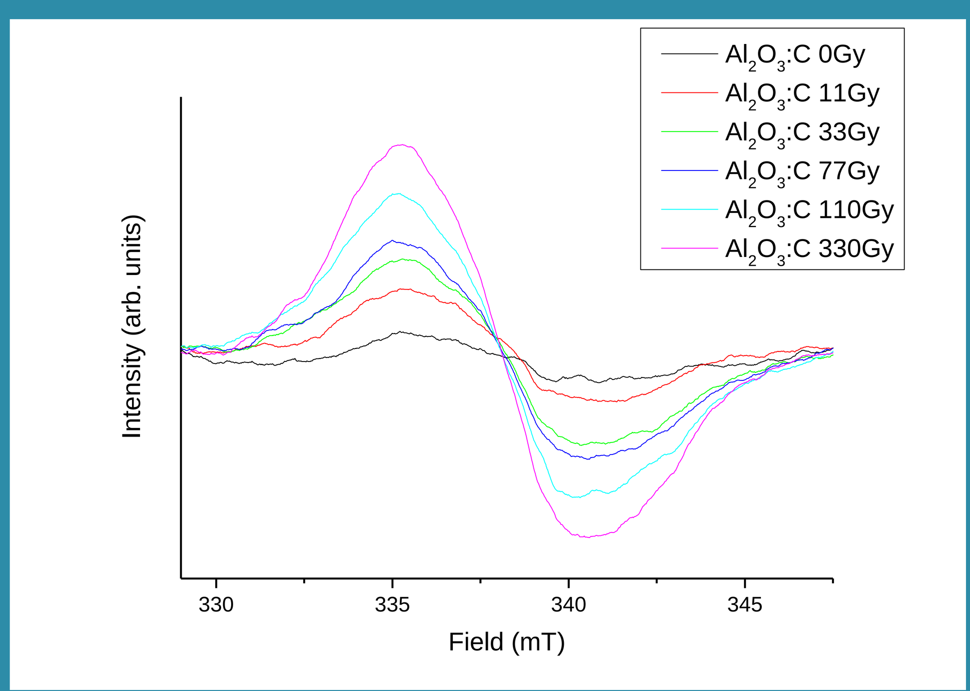
Their intensities are also different. The Al_2O_3:C,Mg EPR signal is smaller by a factor of around 2 for the same dose received.

Dose dependence

Only Al_2O_3:C is shown, Al_2O_3:C,Mg gave similar results.

Looking at the dose dependence there is a clear increase in intensity in function of dose received for both.

X-band EPR dose dependence Al_2O_3:C, RT



Plotting the EPR intensity in function of dose gives an exponential curve from which we could derive an estimate of the saturation dose:

Fit Al_2O_3:C,Mg:

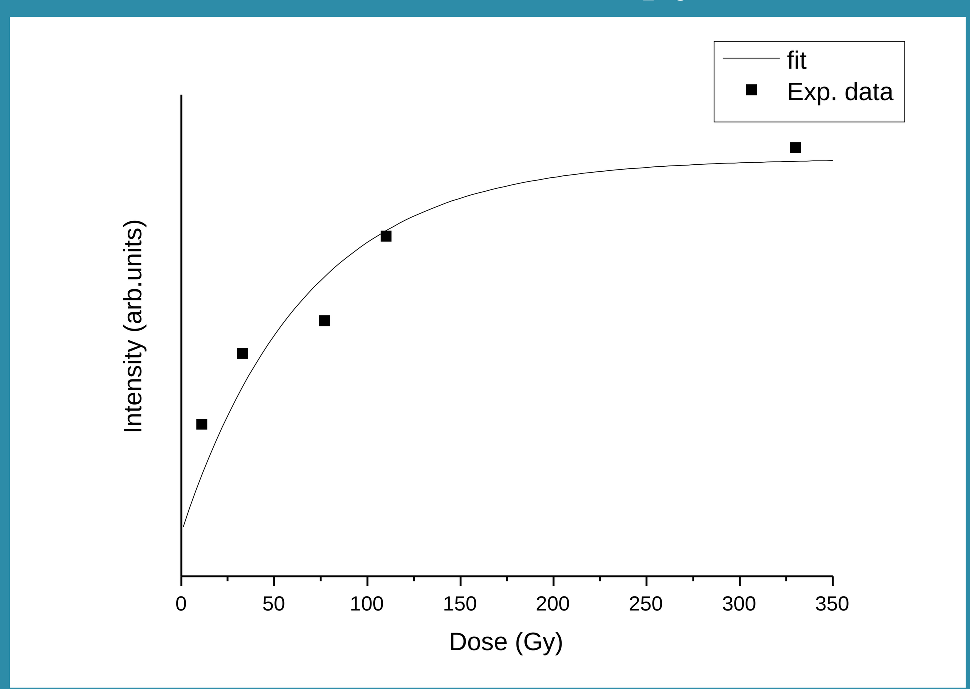
$$I = I_0 * (1 - e^{-\frac{D}{58,8\,Gy}})$$

Fit Al_2O_3:C:

$$I = I_0 * (1 - e^{-\frac{D}{66,7\,Gy}})$$

For both samples the saturation dose is around 60 Gy, close to the OSL/RPL saturation dose.

Fitted dose dependence Al_2O_3:C



Conclusions

- Al_2O_3:C and Al_2O_3:C,Mg have an EPR signal that is dose sensitive
  - The EPR signal of Al_2O_3:C is more sensitive to radiation compared to the EPR signal of Al_2O_3:C,Mg
  - The saturation dose is around 60 Gy, in agreement with OSL saturation dose
- LiF:Mg,Cu,P has an EPR signal present that is not dose sensitive
  - The signal could be related to Cu^{2+}, however more research is needed
  - The first ENDOR spectra look promising

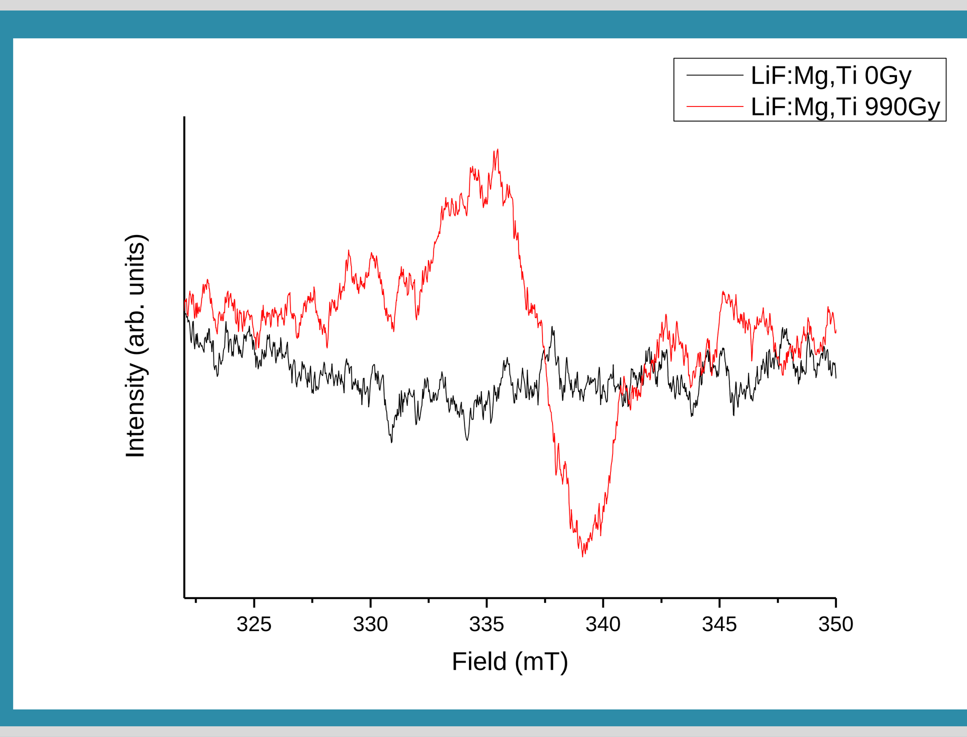
LiF:Mg,Ti

X-Band measurements, RT

Measurements before irradiation did not reveal a significant EPR signal.

After irradiation a signal appears, but even for 1 kGy of dose, the intensity remains very low.

g = 2,014 lwpp = 3,8 mT



LiF:Mg,Cu,P

Q-Band measurements

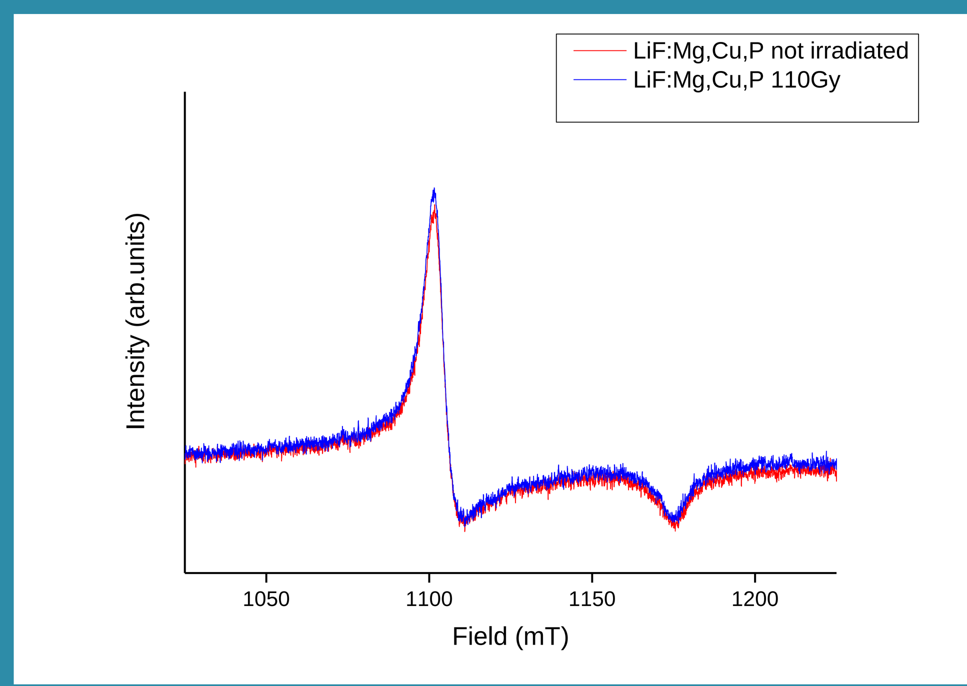
For this system a signal is present before irradiation.

$g_\perp$ = 2,204 $g_{//}$ = 2,068

lw = 8,3 mT (Lorentzian)

Irradiating the sample did not change the intensity of the signal, nor its shape.

Q-Band EPR, RT



Literature

Patil and Moharil 1995 [5]

$g_\perp$ = 2,191 $g_{//}$ = 2,073 (X-band, RT)

The signal is proposed Cu-related.

Prediction g-values Cu^{2+}

Crystal Field Theory	DFT calculations
$d_{x^2-y^2}$: $g_{//} > g_\perp > 2$	$g_\perp$= 2,115 < $g_{//}$=2,259
$d_{3z^2-r^2}$: $g_\perp > g_{//} \approx 2$	$g_\perp$= 2,219 >$g_{//}$=2,005

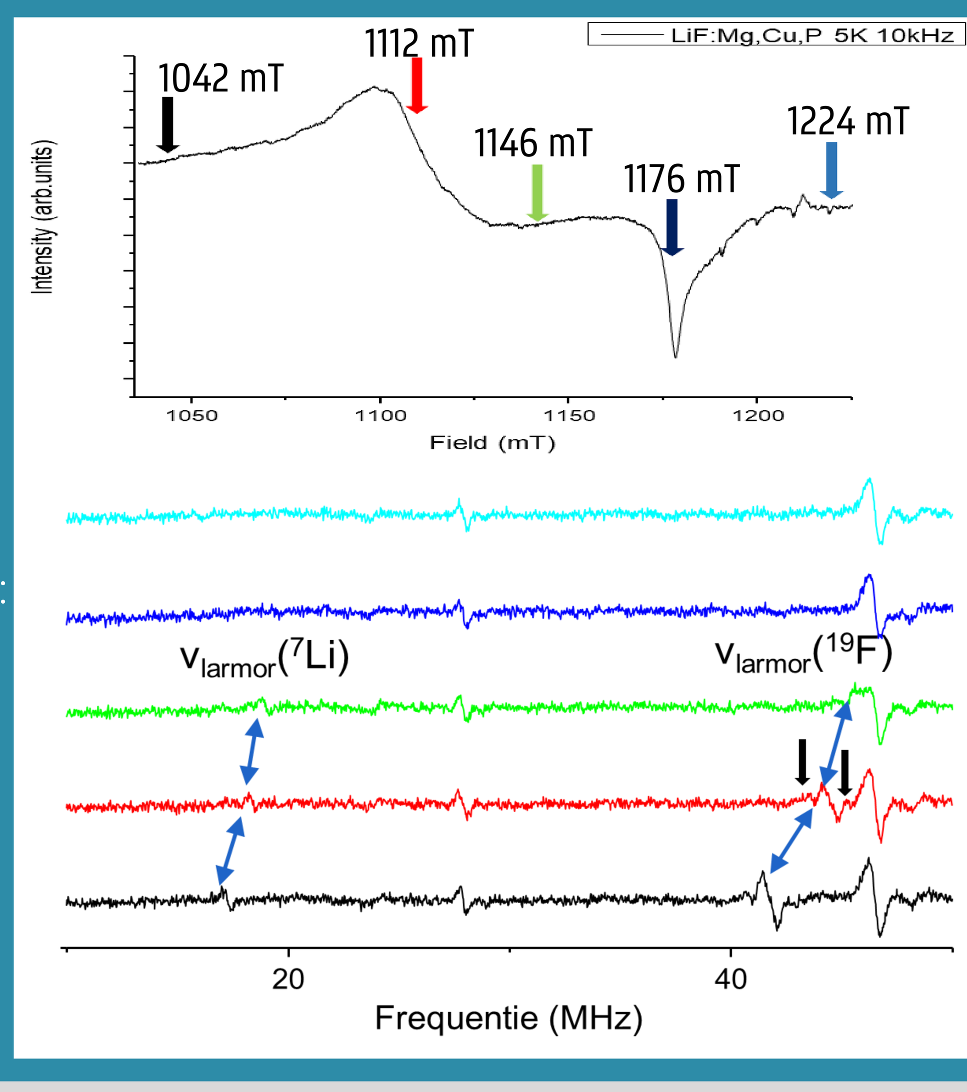
CFT predicts that the unpaired electron is either in $d_{x^2-y^2}$ or in $d_{3z^2-r^2}$ orbital. For these orbitals theoretical g-values can be calculated. DFT calculations confirmed the CFT predictions. However, comparing this to the experimentally derived g-values, neither really fit.

Q-band ENDOR measurements

Electron Nuclear Double Resonance (ENDOR): Detecting NMR transitions via EPR.

First results reveal signals at the Larmor frequencies of ^{7}Li and ^{19}F from distant nuclei. Around $\nu_{larmor}(^{19}F)$ a signal is visible with a hyperfine coupling of 0,9 MHz due to a neighbouring F nucleus.

Q-band EPR + ENDOR, 5K



Acknowledgements

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References

1. M. Ikeya, New applications of electron spin resonance: dating, dosimetry and microscopy. (World Scientific Publishing, 1993).
2. E. G. Yukihara, and Stephen WS McKeever, Optically stimulated luminescene: fundamentals and applications. (John Wiley & Sons, 2011).
3. M. J. Aitken, Thermoluminescence dating. (Academic press, 1985).
4. L. F. Nascimento, M. Karampiperi, J. P. Oliveira and F. Vanhavere, Radiation Measurements (2017).
5. R R Patil and S.V.Moharil, J. Phys.: Condens. Matter 7 (025), 9925-9933 (1995).

Contact

Kwinten Maes
PhD assistant
DEPARTMENT OF SOLID STATE SCIENCE
EMR RESEARCH GROUP
E kwinten.maes@ugent.be
T +32 9 264 43 51
www.ugent.be

Ghent University
@ugent
Ghent University

GHENT UNIVERSITY

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